AD737627

AMERICAN OPTICAL
CORPORATION
CENTRAL RESEARCH LABORATORY • SOUTHBRIDGE, MASS. 01550

16

UNCLASSIFIED

Security Classification

DOCUMENT CONTROL DATA - R & D

(Security classification of title, body of abstract and indexing annotation must be entered when the overall report is classified)

1. ORIGINATING ACTIVITY (Corporate author) American Optical Corporation Central Research Laboratory Southbridge, MA 01550	2a. REPORT SECURITY CLASSIFICATION Unclassified 2b. GROUP N/A

3. REPORT TITLE

NEODYMIUM LASER GLASS IMPROVEMENT PROGRAM

4. DESCRIPTIVE NOTES (Type of report and inclusive dates)

Technical Summary Report No. 7 1 July 1965-31 December 1965

5. AUTHOR(S) (First name, middle initial, last name)

Richard F. Woodcock

6. REPORT DATE	7a. TOTAL NO. OF PAGES	7b. NO. OF REFS
October 1971	16	None

8a. CONTRACT OR GRANT NO. Nonr 3835(00) b. PROJECT NO. 7300 c. d. ARPA Order No. 306-64	9a. ORIGINATOR'S REPORT NUMBER(S) TR-598-7 9b. OTHER REPORT NO(S) (Any other numbers that may be assigned this report) None

10. DISTRIBUTION STATEMENT

Distribution of this report is unlimited

11. SUPPLEMENTARY NOTES	12. SPONSORING MILITARY ACTIVITY
Work is part of Project DEFENDER	Office of Naval Research Washington, D. C.

13. ABSTRACT

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DD FORM 1 NOV 65 1473

UNCLASSIFIED

Security Classification

14. KEY WORDS	LINK A		LINK B		LINK C	
	ROLE	WT	ROLE	WT	ROLE	WT
Lasers						
Glass						
Athermalization						
Index of Refraction						
Stress Optical Coefficients						
Stress Birefringence						

TR 598-7

This research is part of Project DEFENDER under the joint sponsorship of the Advanced Research Projects Agency, the Office of Naval Research and the Department of Defense.

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Technical Summary Report

Number 7

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Dr. Richard F. Woodcock

Contract No. Nonr-3835(00)
Project Code No. 7300
ARPA Order No. 306

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Efforts during this six month period were a continuation of the investigation of athermalized laser glass materials. Equations were derived for change in optical pathlength, which make it possible to identify two conditions for which athermalization is satisfied. The method used for measurement of thermal coefficient of refraction index is described. Thirty-three glasses were measured and results are given. Finally, search for a material with zero stress birefringence continued. A regression analysis of the stress birefringence coefficient as a function of glass composition was performed on 51 glasses and results are tabulated.

FOREWORD

This report has been prepared by the Central Research Laboratory of the American Optical Corporation, Southbridge, Massachusetts under Contract Nonr 3835(00) entitled "Neodymium Laser Glass Improvement Program." The contract is under the sponsorship of the Office of Naval Research and this report covers the six-month period ending 31 December 1965.

Dr. Richard F. Woodcock is Project Manager and Author of this report.

This program is part of project DEFENDER.

This report is unclassified.

NEODYMIUM LASER GLASS IMPROVEMENT PROGRAM

1. INTRODUCTION

This report is a technical summary covering work performed during period 1 July to 31 December 1965 on amendment No. 3 to contract Nonr-3835(00) entitled "Neodymium Laser Glass Improvement Program." This amendment, received 13 July 1965, was for a 12 month extension of effort effective retroactively to the completion date of the previous amendment on 30 April 1965. Effort during this period was a continuation of the investigation of athermalized laser glass material studies which were curtailed in April 1965 when the previous amendment expired.

2. TECHNICAL BACKGROUND

In order to obtain well collimated emission from laser devices it is not only necessary to start with good optical quality glass, but also to prevent the generation of optical distortion in the cavity due to thermal gradients induced by the pumping process. The goal of the present research is to develop a laser glass composition in which, for a given cavity configuration, optical changes produced by the temperature dependent parameters will nullify each other, thus resulting in an athermalized laser device.

Equations are derived for the change in optical pathlenth as a function of the distance from the axis of the laser rod for both radially polarized and tangentially polarized light rays passing through the laser rod. From these two expressions for the change in optical pathlength it is possible to identify two conditions under which the requirements for athermalization are satisfied.

In the first case for which the stress birefringence is zero, i.e. the stress optical coefficient for light polarized perpendicular to the direction of stress ($B_\perp$) equals the stress optical coefficient for light polarized parallel to the direction of stress ($B_\parallel$), the conditions for athermalization are satisfied by the expression

$$n\alpha_n + \frac{\alpha E}{1-\sigma}(2B) = 0 \quad , \tag{1}$$

where n is the index of refraction, α_n is the thermal coefficient of refractive index, α is the expansion coefficient, E is Young's Modulus, σ is Poisson's Ratio and B is the stress optical coefficient for either direction of polarization. To satisfy this condition, α_n must have a negative value since the other parameters all have positive values and σ is less than unity.

In the second case, where $B_\perp$ and $B_\parallel$ are not equal, the glass exhibits stress birefringence and it is no longer possible to chose a set of parameters which will make the change in pathlength equal to zero for both tangentially and radially polarized light. To satisfy the conditions for athermalization in this case, one can design a cavity such that the direction of polarization of the rays oscillating within the cavity alternates between radial and tangential for each successive transit through a given laser element. This may be achieved by placing an appropriate Faraday rotator between the laser rod and the totally reflecting mirror in a single rod system or between successive laser elements in a cascaded system. Material parameters required to satisfy the conditions for athermalization in this case are given by the expression

$$2n\alpha_n + \frac{\alpha E}{1-\sigma}(B_\perp + 3B_\parallel) = 0 \quad . \tag{2}$$

The various material parameters in these equations are being investigated as a function of laser glass composition to provide information which will make it possible to design a glass composition meeting the above specifications for an athermal laser system.

3. THERMAL COEFFICIENT OF REFRACTIVE INDEX

The method used for the measurement of the thermal coefficient of refractive index, α_n, is one based on the fact that the output spectrum of a laser rod in a laser cavity using a thin plate of transparent material as the output reflector, is controlled by the optical thickness of that thin plate. This occurs because the reflectivity of the plate increases from a nominal 4% per surface in the case of glass to about 16% at those wavelengths for which the optical thickness of the plate is equal to an odd number of 1/4 wavelengths, i.e.

$$nL = (2N + 1)\,\lambda/4 \quad , \tag{3}$$

where n = the index of refraction, L is the plate thickness, N is an integer and the resulting λ's are possible laser emission wavelengths within the fluorescent emission bandwidth. When the temperature is changed, both the physical thickness and the index of refraction of the glass plate will change and thus the wavelength at which laser action may take place will shift. This shift is governed by the following expression which is obtained from Eq. (3), namely

$$\frac{dL}{LdT} + \frac{dn}{ndT} = \frac{d\lambda}{\lambda dT} \tag{4}$$

or

$$\alpha + \alpha_n = \alpha_T \quad , \tag{5}$$

where α is the conventional expansion coefficient, α_n is the thermal coefficient of refractive index and α_T is the coefficient of the shift in wavelength of the laser emission as a function of temperature.

The apparatus for measuring α_T is shown in Figure 1.

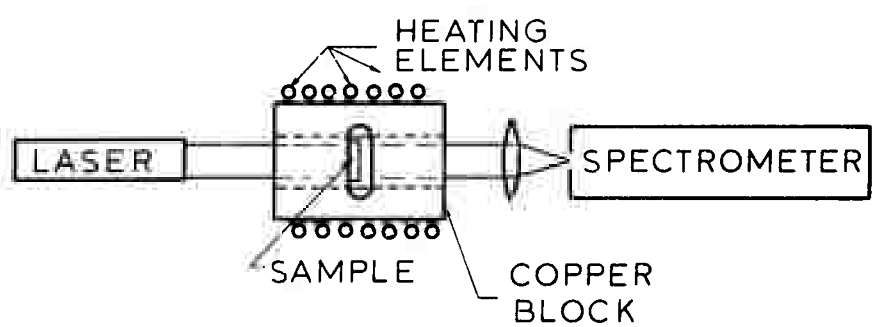

Figure 1. Schematic diagram of apparatus for measuring thermal coefficient of the index of refraction.

A neodymium laser is used in the experiment because we wish to measure α_n at the 1.06 μm wavelength. The material to be measured is fabricated into a plate about 0.5 mm thick with the surfaces of the plate polished to an optical parallelism of about 5 fringes or less per centimeter. This plate which serves as the front reflector of the laser rod is placed in a furnace fabricated from

a solid block of copper to provide thermal stability. Light from this laser system is focused upon the slits of a model No. 70-32 Jarrell-Ash grating spectrometer and successive exposures are made on infrared sensitive Kodak type 1-Z spectroscopic plates at several different temperatures in the range from room temperature to 100° C. The plate is moved vertically between exposures.

Spectra of a typical glass sample as a function of temperature are shown in Figure 2.

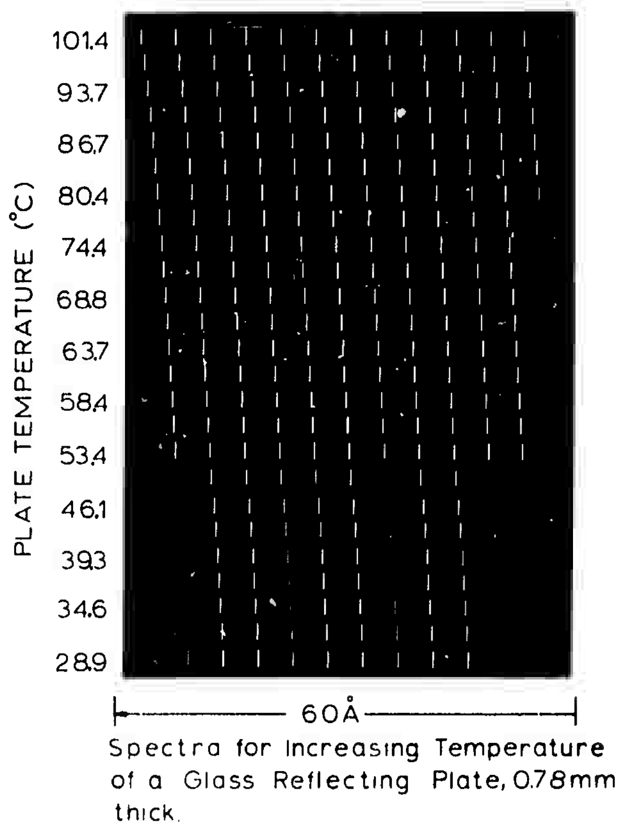

Figure 2. Spectral output of a glass laser with a thin plate acting as a reflection filter for various temperatures of the plate.

The shift in emission spectra is obtained from these spectra and from this data a curve of $\Delta\lambda/\lambda$ vs T may be plotted, the slope of which is α_T. These plots are not always straight lines because the expansion coefficient, α, is not a true constant as a function of temperature. By making a separate measurement of the thermal expansion coefficient and plotting $\Delta L/L$ vs temperature

on this same graph, as shown in Figure 3, it is possible to take the difference between these two curves on a point by point basis and from a plot of these difference values to determine the curve for $\Delta n/n$ as a function of temperature, the slope of which is α_n.

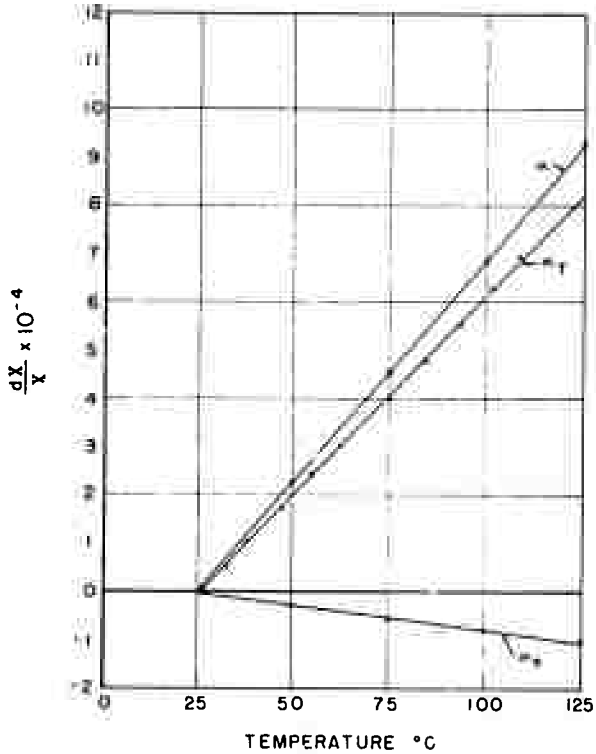

Figure 3. Effect of temperature on the parameter X where X may be length (L), emission wavelength (λ) or refractive index (n). The slopes of the curve are α, α_T and α_n respectively. The curve for α_n is derived from the other two curves.

This measurement has now been completed for a series of 33 glasses, the compositions of which were designed to include a wide variation in batch ingredients and their concentrations, in order to provide information for the design of optimized glass compositions. Results of these measurements are given in Table I. In order to satisfy the conditions for athermalization set forth in either Eqs. (1) or (2), α_n must be a negative value, as previously stated, and the ratio of α_n/α, a preliminary figure of merit for athermalization, should probably be as large as possible. Values of this figure of merit are also included in Table I.

TABLE I

THERMAL COEFFICIENT OF REFRACTIVE INDEX, α_n, AND PRELIMINARY ATHERMALIZATION FIGURE-OF-MERIT α_n/α

GLASS NUMBER	α_n ($\times 10^{-7}$)	α_n/α
1175	35	0.42
1191	- 5	-0.05
1192	3	0.04
1193	13	0.19
1194	5	0.04
1195	8	0.09
1196	- 9	-0.10
1197	-15	-0.16
1199	-16	-0.13
1200	-13	-0.13
1201	-16	-0.16
1202	0	0
1203	-15	-0.12
1204	-29	-0.24
1233	-20	-0.15
1260	-16	-0.15
1261	-11	-0.13
1262	-11.5	-0.14
1263	-24.5	-0.22
1264	- 9	-0.09
1266	- 4	-0.04
1267	-12.5	-0.11
1268	10	0.09
1269	8	0.12
1270	-28	-0.24
1271	0	0
1273	-18	-0.18
1274	-12.5	-0.13
1275	0	0
1276	-21	-0.19
1465	- 5.7	-0.06
1467	7	0.08
1468	11	0.15
3835	-11	-0.11

designed solely to provide a variety of glass ingredients, some compositions appear to have better athermal properties than our present commercial laser glass. One of the first promising compositions based on the ratio of α_n/α, namely MG-1204, was remelted with slight adjustments in composition; such as an increase in Nd_2O_3 concentrations, which appeared to be too low for a good laser characteristics. If the athermal properties of this composition, MG-1750, are still encouraging, a larger melt will be made to provide glass of good optical quality in order that a direct measurement can be made of the induced optical distortion produced in a laser rod of this material during the pumping process. A comparison will be made between the distortion induced in this glass and our standard AOlux commercial laser material.

4. STRESS BIREFRINGENCE

Some effort continued in the search for a material with a zero value of stress birefringence, the case for which Eq. (1) defines the conditions for an athermalized system. Stress birefringence is here defined as the difference in the stress optical coefficients for light polarized parallel to and perpendicular to the direction of thrust respectively. This value is commonly referred to as the stress optical coefficient which leads to some confusion in terminology. Following the sign convention of Adams and Williamson and others, it is defined here as $B_{\parallel}-B_{\perp}$.

A regression analysis of the stress birefringence coefficient as a function of glass composition was made resulting in the regression coefficients b_i listed in Table II for the glass ingredients listed therein. These results show some general trend in the effects of composition on stress birefringence but the accuracy of the regression coefficients in general are too poor to give any strong indications of new compositions which might lead to glasses with zero stress birefringence. Lead oxide glasses were not included in this analysis study because glasses of the Pockels type are not particularly good laser materials and have the disadvantage that their thermal coefficient of refractive index is a positive value whereas a negative value is required to satisfy the conditions of athermalization as previously stated. The 51 glasses included in this analysis, their measured stress birefringence coefficients ΔB and the values of stress birefringence calculated from the regression coefficients listed in Table II and tabulated in Table III.

TABLE II

REGRESSION COEFFICIENTS (b_i) OF THE STRESS-BIREFRINGENCE COEFFICIENT (ΔB)

PERIODIC GROUP	GLASS COMPONENT	REGRESSION COEFFICIENT	σ (r.m.s. DEVIATION)	RANGE OF X_i values (wt %)
IA	Li_2O	0.007	0.047	0-7.7
	Na_2O	-0.007	0.020	0-24.1
	K_2O	-0.003	0.020	0-30.1
	Rb_2O	-0.012	0.011	0-40.6
IIA	CaO	-0.001	0.047	0-34.2
	BaO	0.002	0.006	0-51.4
IIB	ZnO	-0.051	0.060	0-9.0
IIIA	La_2O_3	-0.012	0.020	0-22.0
	CeO_2	-0.030	0.109	0-5.0
	Nd_2O_3	-0.063	0.048	0-7.0
IIIB	B_2O_3	-0.012	0.019	0-15.0
	Al_2O_3	-0.068	0.031	0-22.0
IVA	TiO_2	-0.046	0.015	0-42.0
IVB	SiO_2	-0.033	0.007	0-70.4
	SnO	-0.040	0.140	0-3.1
VA	Nb_2O_3	-0.034	0.030	0-15.4
	Ta_2O_5	-0.042	0.022	0-26.3
VB	P_2O_5	-0.013	0.009	0-73.1
	Sb_2O_3	0.086	0.088	0-4.2
	Bi_2O_3	-0.009	0.006	0-85.0

TABLE III

COMPARISON OF MEASURED AND CALCULATED VALUES OF STRESS BIREFRINGENCE

COMPOSITION NUMBER	ΔB (BREWSTERS) MEASURED	ΔB (BREWSTERS) CALCULATED	COMPOSITION NUMBER	ΔB (BREWSTERS) MEASURED	ΔB (BREWSTERS) CALCULATED
MG-256	-2.6	-2.3	MG-1267A	-1.2	-1.7
MG-372	-0.5	-0.6	MG-1268C	-2.0	-1.4
MG-467	-2.6	-2.6	MG-1269A	-2.5	-2.3
MG-702D	-2.9	-2.7	MG-1270A	-2.2	-2.1
MG-1015A	-1.2	-1.2	MG-1271B	-2.2	-2.4
MG-1050A	-3.1	-3.0	MG-1272A	-2.3	-2.1
MG-1191A	-2.2	-2.27	MG-1273A	-1.4	-1.6
MG-1192B	-1.9	-1.7	MG-1274B	-2.2	-2.3
MG-1194A	-2.1	-2.0	MG-1275A	-2.3	-2.2
MG-1195A	-2.4	-2.5	MG-1276A	-2.0	-1.2
MG-1196A	-2.2	-2.1	MG-1465A	-1.9	-1.2
MG-1197A	-1.3	-1.6	MG-1466B	-2.5	-2.3
MG-1200A	-2.3	-2.2	MG-1467A	-2.4	-2.2
MG-1201A	-1.6	-2.3	MG-1468A	-2.3	-2.3
MG-1203A	-2.2	-2.1	MG-1479B	-2.1	-2.2
MG-1204A	-2.2	-1.9	MG-1480B	-0.6	-0.9
MG-1217A	-2.0	-2.3	MG-1482A	-2.2 to -2.4	-2.4
MG-1233B	-1.5	-1.6			
MG-1248B	-2.2	-2.1	MG-1489A	-2.2	-2.0
Mg-1260A	-0.9	-2.0	MG-1504A	-1.5	-1.2
MG-1261B	-2.6	-2.3	MG-1514A	-1.5	-1.3
MG-1262B	-1.6	-1.8	3255	-2.5	-1.9
MG-1263A	-1.6	-2.2	3403	-2.8	-2.2
MG-1264A	-0.7	-2.1	3669	-2.6	-2.6
MG-1266B	-2.8	-1.8	3832	-2.6	-2.4
			3835	-2.4	-2.7
			EB-20B	-2.6	-2.5

The comparison between measured and calculated values was made in an attempt to provide some insight into the poor σ-values in Table II. Considering these σ-values, the agreement between measured and calculated values of ΔB was suprisingly good. It is assumed that the uncertainty in the regression coefficient values is probably due to the fact that for some of these values only a few glasses containing the glass ingredient in question were available for analysis and the range of concentrations used was sometimes rather large. The glasses included in the analysis consisted of the series of glasses designed for the athermalization study, plus a variety of other compositions which would broaden the range of concentration of the glass ingredients included in the study.

A few additional glass compositions were tried during this period in an attempt to fabricate glasses from some of the glass ingredients which gave the most positive values for the regression coefficients as listed in Table I. Not all these compositions made good glass and those which did make good glass with stress birefringence values near zero either contained B_2O_3 which is known to seriously quench the fluorescent lifetime of the Nd^{3+} ion or they were glasses in which the network former was phosphate rather than silicate. This type of host glass has not as yet proven to be a reliable material for laser applications due possibly to greater difficulties in obtaining high purity ingredients and good optical quality glass from the melts.

From these results it would appear that the chances were rather remote for obtaining a satisfactory laser glass material in which the stress birefringence was zero so that the conditions for athermalization would be satisfied by Eq. (1). Future efforts to obtain an athermalized material will therefore be confined to the case in which the stress optical coefficients differ for rays polarized parallel to and perpendicular to the direction of thrust, in which case the conditions for athermalization will be governed by Eq. (2).